Designing a Rieske Route for C-H Bond Functionalization

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One of Nature's underexplored strategies for facilitating C–H bond functionalization involves the class of Rieske Oxygenases. These enzymes use a high valent Fe-based oxidant to facilitate a diverse set of powerful and specific transformations, including monooxygenation, dioxygenation, and sequential monooxygenation reactions. This chemistry is vital to a number of biosynthetic and degradative pathways and thus, these enzymes have been recognized for their potential use in building complex natural products and degrading environmental pollutants. However, the practical applicability of Rieske Oxygenases is limited by a gap in knowledge regarding the structure–function relationships in this class of enzymes. Here, I will detail our progress towards identifying the architectural motifs that Rieske Oxygenases employ to dictate site-selectivity, substrate specificity, and reaction outcome. Collectively, this work provides a framework for structurally reprogramming a Rieske oxygenase.